

Thermoelectric properties of disordered systems

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1. Introduction

The electronic properties of disordered systems have been the subject of intense study for several decades. Thermoelectric properties, such as thermopower and thermal conductivity, have been relatively neglected. A long standing problem is represented by the sign of the thermoelectric power.¹⁾ In crystalline semiconductors this is related to the sign of the majority carriers, but in non-crystalline systems it is commonly observed to change sign at low temperatures. In spite of its apparent universality this change has been interpreted in a variety of ways in different systems. We have developed a Green's function recursion algorithm¹⁰⁾ based on the Chester-Thellung-Kubo-Greenwood formula²⁻⁴⁾ for calculating the kinetic coefficients L_{ij} on long strips or bars. From these we can deduce the electrical conductivity σ , the Seebeck and Peltier coefficients S & Π and the thermal conductivity κ , as well as the Lorenz number L_0 . We present initial results for 1D systems. In 1D we observe a Lorentzian-like distribution for the thermopower which is modified by the presence of inelastic scattering. This could give rise to non-negligible quantum fluctuations in macroscopic systems at low temperatures.

Within the linear response approach, the responses of a system to an external electric field \mathbf{E} and a temperature gradient ∇T up to linear order⁵⁾ are

$$\langle \mathbf{j} \rangle = |e|^{-1} (|e|L_{11}\mathbf{E} - L_{12}T^{-1}\nabla T) \\ = \sigma\mathbf{E} - \sigma S\nabla T \quad (1)$$

$$\langle \mathbf{j}_q \rangle = e^{-2} (|e|L_{21}\mathbf{E} - L_{22}T^{-1}\nabla T) \\ = ST\langle \mathbf{j} \rangle - K\nabla T. \quad (2)$$

Here, $\langle \mathbf{j} \rangle$ and $\langle \mathbf{j}_q \rangle$ denote charge and heat currents, L_{ij} , $i, j = 1, 2$ are the kinetic coefficients and e , T , σ , S , and K are electron charge, temperature, conductivity, thermopower and heat conductivity, respectively. Other commonly considered thermoelectric transport coefficients are the Peltier coefficient, Π , and the Lorenz number, L_0

$$\Pi = \frac{L_{12}}{|e|L_{11}} = ST \quad (3)$$

$$L_0 = \frac{L_{22}L_{11} - L_{21}L_{12}}{(k_B T L_{11})^2} = \frac{e^2}{k_B^2} \frac{K}{\sigma T}, \quad (4)$$

with k_B the Boltzmann constant. Calculations of the thermoelectric transport properties that have been done previously are based either on the CTKG formulation²⁻⁴⁾ or other perturbative methods.^{6,7)} These methods are macroscopic in nature and rely on the explicit, phenomenological form $\sigma(E_F, T)$. A microscopic approach to determine the thermoelectric transport properties without assuming explicitly a functional form of σ would be quite instructive. To our knowledge, there has only been Ref.⁸⁾ which reported on the distribution of S in a 1D disordered wire in the localized regime. The authors were able to analytically and numerically show that in this case the distribution of S is Lorentzian. So far there has been no other microscopic calculation of S , Π , K or L_0 in disordered systems near the MIT.

2. The recursive Green function method

This method was used previously to calculate the dc and ac conductivity tensors and the density of states of a d -dimensional disordered system^{9,10)} with and without a magnetic field. Here we will show briefly how the method can be adopted to calculate all kinetic coefficients and thus the other transport properties.

The 1D Hamiltonian can be written as

$$H = \sum_{j=1}^N \epsilon_j c_j^\dagger c_j + c_j^\dagger c_{j+1} + c_{j+1}^\dagger c_j, \quad (5)$$

where the disorder terms $\epsilon_j \in [-W/2, W/2]$ are randomly distributed. In adding one more site to the chain, the Hamiltonian can then be considered as an unperturbed Hamiltonian of the first $N - 1$ sites and the decoupled N^{th} site plus a perturbation.⁹⁾ Equation (5) then simplifies to

$$H^{(N)} = [H^{(N-1)} + \epsilon_N c_N^\dagger c_N] + [c_{N-1}^\dagger c_N + c_N^\dagger c_{N-1}] \quad (6)$$

The last part in the Hamiltonian, H' , is the coupling between the $(N - 1)^{\text{th}}$ site and the N^{th} site. This coupling can be treated iteratively using the one-particle Green function $G^\pm(z^\pm)$ as

$$G^{(N)} = G_0^{(N-1)} + G_0^{(N-1)} H' G^{(N)} \quad (7)$$

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where $z^\pm = E \pm i\xi$, where E is the energy and ξ may be interpreted as an inverse lifetime, e.g. due to inelastic scattering.

3. Results and Conclusions

For brevity, we shall restrict ourselves here to studying the behavior of $S(L_{12})$. In complete analogy to the formulation for $\sigma (= L_{11})$,^{9, 10)} we can compute $L_{12}(T = 0)$ using the recursive equations

$$\begin{aligned} L_{12}^{(N)} &= (E - \mu)\sigma^{(N)} - \xi \frac{e^2}{\pi \hbar N a} \text{Im} f_N^+, \\ h_N^+ &= g_N^{+2} [(x_N - x_{N-1})^2 g_{N-1}^+ + h_{N-1}^+] , \\ f_N^+ &= f_{N-1}^+ - 2g_N^+ [(x_N - x_{N-1})^2 g_{N-1}^+ + h_{N-1}^+] . \end{aligned} \quad (8)$$

The initial condition is $f_0^+ = h_0^+ = 0$. From Eq. (3) we see that

$$\Pi^{(N)} = \frac{1}{|e|} \left\{ (E - \mu) - \frac{\xi \text{Im} f_N^+}{4 s_N} \right\} \propto L_{12}^{(N)} . \quad (9)$$

where $s_N = \pi \hbar N a \sigma^{(N)}/4e^2$. The distribution of L_{12} (in units of $e^2/\pi \hbar a$) is shown in Fig. 1.

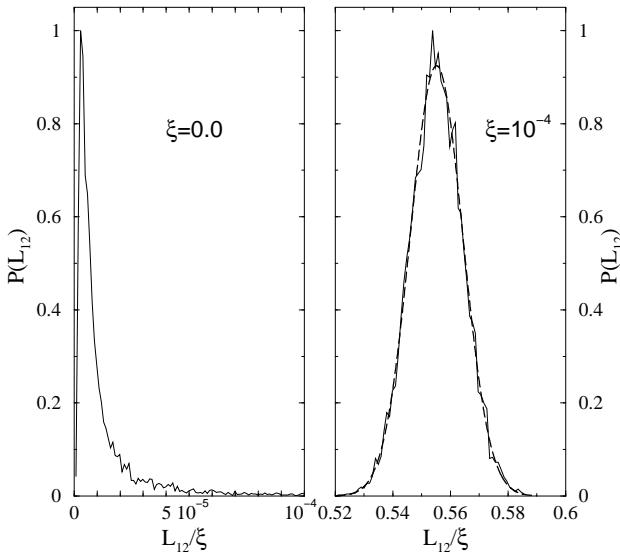


Fig. 1. Distribution function $P(L_{12})$ for zero and finite value of ξ at $E = \mu = -1$, $W = 2$ and $T = 0$. Data is computed from 10000 samples and length $Na = 3 \times 10^6 a$. P is normalized to 1 at the maximum. The dashed line correspond to a Gaussian (right) fit.

A further interesting quantity is the *dimensionless thermopower* τ defined via

$$\tau = \frac{\Delta}{2\pi g(E)} \frac{dg(E)}{dE} \Big|_{E=E_F} = \frac{\Delta}{2\pi} \frac{d}{dE} \ln |G_{1N}^+(E)|^2 , \quad (10)$$

with Δ the mean level spacing near E_F and g the conductance. In the limit of weak scattering the dominant effect of the disorder is simply to introduce a finite mean free path at energies well inside the band. This implies that on the average (10) can be approximated by $\tau = -\frac{\Delta}{\pi} \text{Tr} [\text{Re} G^+(E)]$. The full distribution can be shown to have a simple Lorentzian form⁸⁾ which can be

compared with the results of the recursive approach as shown in Fig. 2.

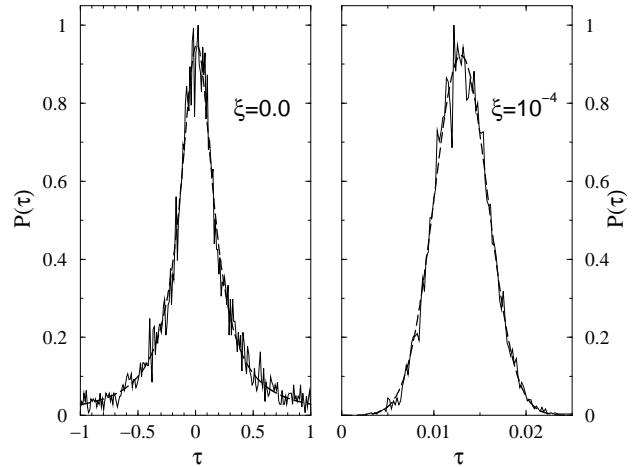


Fig. 2. Distribution function $P(\tau)$ for zero and finite value of ξ and the same parameters as Fig. 1. The dashed lines correspond to a Lorentzian (left) and a Gaussian (right) fit.

Using $O(N)$ arithmetic operations, where N is the number of sites in the system, we can compute iteratively all kinetic transport coefficients L_{ij} . The maximum value of N is mainly limited by the computer time available and the intended precision of the results. In this short note, we have considered only the transport properties of a chain initially having N sites of a 1D tight-binding model with nearest neighbor hopping in the limit $T = 0$. It is straightforward to extend the computations to higher dimensions¹¹⁾ and results will be presented elsewhere. Detailed derivations of relevant equations and recursion relations are given elsewhere.¹²⁾

We have shown that the distribution of the dimensionless thermopower τ is indeed Lorentzian as predicted⁸⁾ but that this changes to Gaussian on introduction of a finite lifetime. Similarly the distribution of L_{12} is Lorentzian-like (albeit asymmetric) for $\xi \rightarrow 0$ but also becomes Gaussian for finite ξ .

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